

The Impact of Long-Term Fertilization on Nitrogen Leaching from Double Rice Field of Hunan, China

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Abstract

To investigate the impact of long-term (1981-2007) fertilization on nitrogen leaching in soil solution of double rice field, a two years' study was conducted in Hunan, China. Porous suction pipes was employed to extract soil solutions from different aquifers (in depth of 20-30 cm, 50-60 cm and 80-90 cm) of rice field to analyze the concentration of ammonium-N ($\text{NH}_4^+\text{-N}$) and nitrate-N ($\text{NO}_3^-\text{-N}$) and the total loss of nitrogen. Results showed that nitrate-N was the primary form of N leaching in studied area. Significant seasonal variance as well vertical distribution of N leaching was found. $\text{NH}_4^+\text{-N}$ in soil leachate mainly occurs in submerged situation before mid-season drainage, while $\text{NO}_3^-\text{-N}$ could primarily be found in situation of saturation especially in late growing stage. Concentration of $\text{NH}_4^+\text{-N}$ decreased with the increasing sampling depth in range of 0-100 cm; whereas for the $\text{NO}_3^-\text{-N}$, the vertical distribution was complicated: it increased in 0-60 cm and then decreased in 60-90 cm. Long-term incorporation with rice straw (NPKS) causes greater total N losses than applied chemical fertilizer only (NPK), and N leaching of CK (No fertilizer) was significantly less than these two treatments. For the studied area, comparatively higher N leaching losses occurred due to long-term fertilization, but with low potential hazards for nitrate contamination due to no extreme high $\text{NO}_3^-\text{-N}$ was found in all samples.

Introduction

Fertilization is the primary non-point source of nitrate leaching in groundwater (Hubbard and Sheridan 1994; Schilling and Wolter 2001). In irrigated lands, rice (*Oryza sativa* L.) field accounts for a pronounced part of fertilizer consumption among agricultural crops while urea is

the most commonly used N fertilizer (Chowdary et al. 2004). Actually, nitrogen (N) represents approximately 70 % of the total nutrient consumption in Asia and almost 60 % of the N fertilizer is used in rice field (Stangel and Datta 1985). China has the world's second largest rice growing area (31.3 million ha) (Ghosh and Bhat 1998) but has the largest rice production (1.66×10^8 T in 2003) (FAO 2004). As the staple food, rice production is the most important factor for Chinese food structure. However, researches over the past decades showed that N fertilizer in rice field was generally inefficient, with N use efficiency less than 40 % of the applied N, therefore, mitigating chemical N fertilizer emission to soil or groundwater remain an critical goal of effective N-management for the sustainability of high yields of wetland rice (Singh et al. 2001).

In fact, the application rates of N-fertilizers were increasing in last two decades and the NO_3^- -N contamination of agricultural water was ascending in many areas of China (Zhu et al. 2000). Some studies employed the lysimeters and indoor's emulation method to observe N leaching in groundwater from rice field of China (Zeng and Wu 2007; Wang et al. 1996; Wang et al. 2009). Nevertheless, there was almost no study focused on influence of long-term fertilization on N leaching in double rice field of China. Both fertilizer type and application rate could affect the N leaching in soil leachate. Wang et al. (2009) found equivalent ammonium bicarbonate and urea can lead marked difference in nitrate and ammonium leaching. Zeng et al. (2007) also concluded there was a notable positive correlation between N application rate and total N losses. By another study in Hunan, China, the controlled release N fertilizer resulted in less nitrate-N loss in groundwater of rice paddy compared to equivalent urea (Ji et al. 2007). In addition, incorporated organic manure leads a greater nitrate loss compared to only using chemical N (Tong et al. 1997). Furthermore, the application times of fertilizer may lead a different leaching losses of N. Zhu et al. (2000) and Tong et al. (1997) considered that splitting N application into several times could decrease the N leaching losses. Besides fertilization, water management regime can cause a pronounced effect on N leaching in leachate of rice field. Pande and Adak (1971) found in submerged rice field, NH_4^+ -N dominated the N leaching, while under saturation, nitrate led the fashion. Sahu and Samant (2006) also considered that the high level of the percolation of soil water caused high leaching losses of N. In another study, Lu et al. (2006) found in continuous submergence rice field, nitrate leaching was well suppressed. Nevertheless, the knowledge of interactions between water regime and fertilization on N leaching in soil solutions of double rice field still not well described.

Suction-cup method has been used to extract soil solutions in cropland over years (Goulding and Webster 1992; Lord and Shepherd 1993). The advantages of suction cups are their simple installation and the negligible disturbance of the soil profile (Grossmann and Udluft 1991), Webster et al. (1993) also indicated the suction cups provided an accurate measure of N leaching losses from arable land. However, little information is available on the effect of long-term fertilization on soil water N leaching from Chinese double rice field. Hence the present investigation was conducted to investigate the impact of long-term fertilization on NH_4^+ -N and NO_3^- -N leaching losses in double rice field of Hunan, China, using the porous pipes based on the suction-cup method.

Materials and methods

Site information and Long-term fertilization design

The long-term fertilization experiment site (Key field monitoring experimental station for reddish paddy soil eco-environment in Wangcheng, Ministry of Agriculture of China) located in Wangcheng County, Hunan Province, China ($112^{\circ}36' \sim 113^{\circ}2'E$, $27^{\circ}58' \sim 28^{\circ}34'N$). Figure 1 shows the location of the long-term fertilization experiment station. The site has an annual mean temperature of 17.5 centigrade, with an accumulative temperature ≥ 10 centigrade for 5450 centigrade d^{-1} , an annual sunshine of 1700 h, and annual precipitation ranging between 1300~1400 mm. The latosolic red soil under experiment was developed from the quaternary period, presents a light dust property. In 1980, before the long-term fertilization experiment started, the physicochemical characteristics of the soil are: pH 6.6, organic carbon 20.6 g kg^{-1} , nitrogen 2.05 g kg^{-1} , and alkali dispelled nitrogen $151.00 \text{ mg kg}^{-1}$. Rice cultivars in early and late rice growing season of 2006 were: Jin You-974 and Xin Xiangyou-80, and for 2007 they were: Zhong Zu-1 and T You-207. The rotation regime was early rice (April-July)-late rice (July-October)-winter fallow (October-April). Water regime of this site was densely intermittent irrigation plus mid-season drainage (MSD) and sparse irrigation (after MSD).

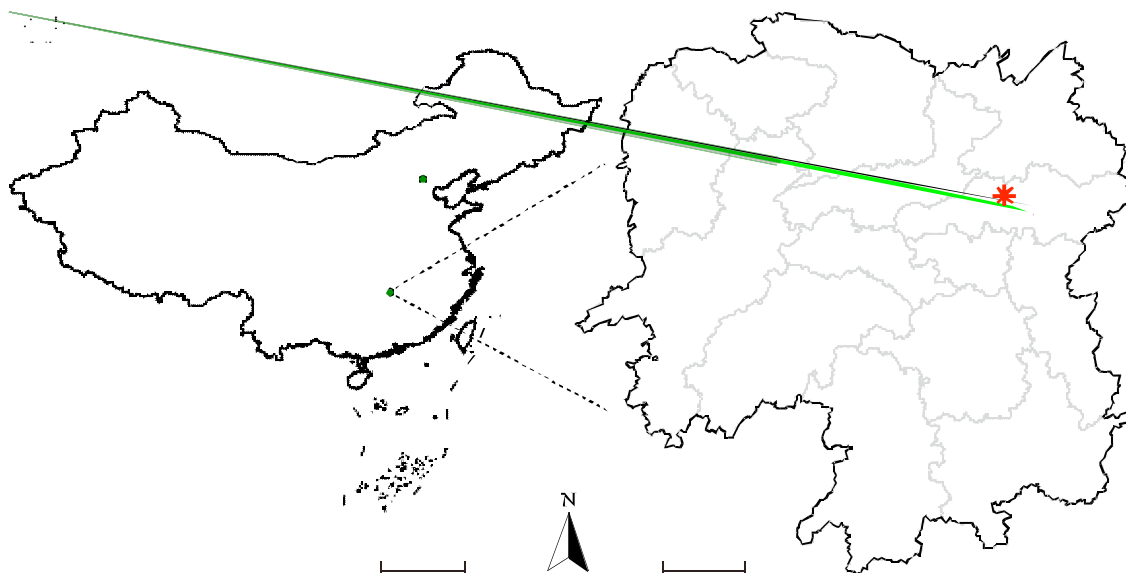


Figure 1. Geographical location of the long-term fertilization experiment station

Since 1981, the long-term fertilizer monitor station has kept nine different fertilizer treatments. Three of them have been chosen to observe nitrate and ammonium leaching from 2006 to 2007: 1) NPKS (Urea, Phosphorus (P_2O_5), Potassium (K_2O), rice straw (S)); 2) NPK (Urea, Phosphorus (P_2O_5), Potassium (K_2O)); 3) CK (No fertilizer). Area of each plot is 66.7 m^2 with

randomized block design and three duplicates. The rate of nitrogen is 150 kg N ha⁻¹ for early rice growing season and 180 kg N ha⁻¹ for late rice growing season. The application of N fertilizer in each season was split into two parts: the first half was used as basal fertilizer just 1 d before transplanting and the other half was applied about 10 d after that as top-dressing. Rice straw applied was the straw from the other plot in the same long-term fertilizer station. The long-term fertilizer design was shown in Table 1.

Table 1. Designs of Long-Term Fertilization (kg ha⁻¹)

	Basal fertilizer				Top-dressing
	N	P ₂ O ₅	K ₂ O	Rice straw	N
CK	0	0	0	0	0
NPKS	75/90*	45	120	2625	75/90
NPK	75/90	45	120		75/90

*75/90 means N applied at early and late rice growing season respectively

Porous suction cups

The approach of the porous suction cups to sample the soil solution was first introduced by Briggs & McCall (1904), due to its relatively simple and cheap to use, this technique has been widely used and modified by numerous researchers since then (Wood 1973; Hansen and Harris 1975). By Grossmann and Udluft (1991), the suction cups consist of hydrophilic materials with fine pores, a portable pump or a syringe is enough to form the suction environment, after the suction is engendered in the sampling system, water is sucked inwards out of the pores of the cup until a corresponding capillary pressure appears in the pores (Grossmann and Udluft 1991). Soil solution then flows from the soil into the cup if the capillary pressure in the cup is lower than that in the soil and will not stop until the capillary pressure is equal inside and outside the suction cup.

Comparing to the soil sampling, this kind of direct solution sampling method has some advantages, such as higher sensitivity for soil mineral nitrogen (Lord and Shepherd 1993), simple to install and minimal soil disturbance (Grossmann and Udluft 1991). The most frequently used suction cups are ceramic, while many other kinds of cups have been reviewed by Grossmann et al. (1987), PVC, PP, PVDF and Teflon were all included. Moreover, the suction cups made of plastic materials can minimize the sorption effects of nitrate-N in soil solution, and many studies showed that plastic cups have some advantages over conventional cups (Grossmann and Udluft 1991).

However, questions about validities of the suction-cup method still remain, due to the difficulty of measurement of leaching losses of mineral-N in cropland (Ramos and Kücke 2001). In fact, both the reliability and the results of the method can be influenced by the operations during the whole sampling procedure (Lord and Shepherd 1993). These operations included which should be noted are preparation of the sampling system (Grossmann and Udluft 1991), installation of the suction cups (depth and angle of probes) (Ramos and Kücke 2001; Webster et al. 1993; Grossmann and Udluft 1991; Lord and Shepherd 1993; Poss et al. 1995), details of sampling

(frequency of sampling) (Webster et al. 1993; Lord and Shepherd 1993) and variability of the properties investigated (Grossmann and Udluft 1991; Webster et al. 1993).

Soil solutions extraction and analysis

Porous pipes used were made of PVC (polyvinyl chloride). The pipes were 3.5 cm in diameter, contains about 300 pores, 0.5 mm in diameter, and 10 cm length to form a porous zone at the end of the pipe with 3 cm margin. The porous zone was wrapped with 4 layers nylon textiles, for which consists of 400 screen meshes in order to prevent big particle impurities entering the pipe. The new porous pipes were cleaned before use by flushing them with dilute acid (Litaor 1988), and before installation, all the pipes were rinsed with a solution similar to the expected soil solution from the site for avoiding the adsorption effects, which new suction cups will exhibit during the first sampling (Grossmann and Udluft 1991).

In April 2006, three depths (30cm, 60cm and 90cm) of holes were drilled by means of the soil auger in each plot before flooding. The slurry of the material from the auger is made and put back into the hole before the probe is inserted for good hydraulic contact between the porous pipe and the soil (Alberts et al. 1977; Barbee and Brown 1986). The pipes were then vertically installed into the holes. Totally 27 porous pipes were installed. All the pipes were then kept in the plot until the harvest of late rice growing season of 2007.

Soil solutions were sampled 7 times in each rice growing season from 2006 to 2007. The specific sampling day depends on rice growing stage and drainage schedule: 1 d, 3 d and 7 d after basal fertilization; before, during and after midseason drainage, and the last time was the day before harvest. The time to extract soil solution was set at 10:00~11:00. Soil solutions in the porous pipe were pumped off four hours before sampling. The pipe was then vacuumed and waited for 4 hours to allow soil solution to enter the pipe. Soil solution of newly entering the pipe was collected by suction pump. The soil solution extraction system is shown in figure 2.

Concentration of NH_4^+ -N in soil solution was analyzed by colorimetric indophenol blue method (Ivancic and Degobbis 1984), and NO_3^- -N density in soil leachate was determined by ultraviolet spectrophotometry (Cawse 1967). The average of seasonal N concentration in each treatment and aquifer was calculated by definite integral depends on its fitting curve of the seasonal variation. The water leaching rate is $3 \text{ mm} \cdot \text{d}^{-1}$ computed by irrigation frequency and area of each plot. Total N losses ratio was calculated using the value of fertilizer N input divided by summary of losses of nitrate and ammonium.

Statistical analysis

Treatment differences in concentration of NH_4^+ -N and NO_3^- -N in two years' four rice growing season were analyzed using SAS PROC MIXED (SAS Institute Inc, 1999) with restricted maximum likelihood option and repeated measures with the compound symmetry covariance structure (Littell et al. 1998). Degrees of freedom were calculated by Kenward-Roger method (Littell et al. 1996). Treatments, season, and their interactions were tested as fixed effects. Means were separated by Fisher's protected LSD test at $P < 0.05$ (Steel and Torrie 1980) and Tukey-Kramer method was used for the P value adjustment.

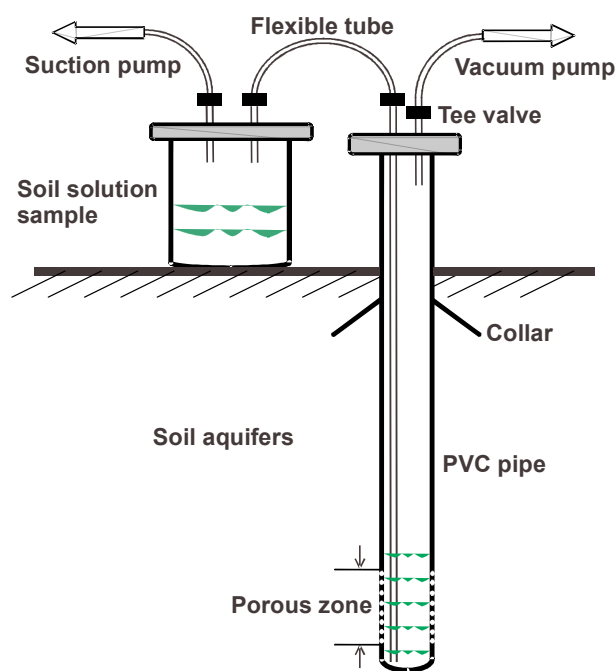


Figure 2. Soil solution extraction system

Results and discussion

Temporal variation of N leaching

Under the impact of long-term fertilization, there was a pronounced seasonal variation of the NH_4^+ -N leaching in NPKS and NPK treatment whereas CK kept a gently low value during the two years' observation, except late rice growing season of 2007 (Figure 3). Statistical analysis indicated that there was significant influence of sampling day on N leaching ($p < 0.01$). In 2006 and early rice growing season of 2007, the NH_4^+ -N concentration in soil leachate was significantly concentrated in first two weeks after basal fertilizer, and then, sharply decreased until harvest, while the top-dressing has almost no effect on this trend. This showed that split application of fertilizer could reduce N leaching in rice field. Some other studies also considered the splitted fertilization could mitigate the N leaching in crop field (Pande and Adak 1971; Arora and Juo 1982). This also indicated that most of the NH_4^+ -N leaching occurred during densely submerged situation before MSD. Wang et al. (2009) considered that the continuous flooding may lead to more NH_4^+ -N leaching than controlled irrigation. In another study, Pande and Adak (1971) believed that NH_4^+ -N losses dominated the N leaching in continuous submerged rice field compared to saturated water regime. Moreover, Liu and Zhang (1999) found NH_4^+ -N is the main form of N in soil solution of submerged rice field due to the long-term densely anaerobic environment, in this kind of situation, the process of ammonification and denitrification was strengthened (McClain et al. 1994), while after MSD, soil moisture and water scheme was totally

different, thus the concentration of $\text{NH}_4^+\text{-N}$ decreased quickly. The summit of ammonium after MSD in late rice growing season of 2007 indicated that there was annual variation in $\text{NH}_4^+\text{-N}$ leaching may be due to different rice cultivars in the two years.

Remarkable temporal variance was also observed in concentration of $\text{NO}_3^-\text{-N}$ in percolation for three treatments (Figure 4). However, the regularity was different from what has been investigated in $\text{NH}_4^+\text{-N}$. Similar seasonal variation curve occurred in CK with NPKS and NPK in 2006 before MSD. In early rice growing season 2006, concentration of $\text{NO}_3^-\text{-N}$ was very high within 7 days after basal fertilizer whereas sharply decreased to nearly zero in following days; but the other peak of nitrate occurred after MSD from 29 DAF (Days after fertilization) to 74 DAF. However, in later rice growing season of 2006, relatively low nitrate-N lasted from 1 DAF to 40 DAF, while a peak was discernible from 40 DAF to 70 DAF. Nevertheless, different situation occurred in 2007, except CK, $\text{NO}_3^-\text{-N}$ in NPKS and NPK expressed similar seasonal variation during early and late rice growing season: a gentle decrease occurred after 1 DAF whereas a big peak was observed until shortly after MSD at 27 and 32 DAF in early and late rice growing season respectively; after MSD, the concentration of nitrate-N declined until harvest in two seasons. The different seasonal variation of $\text{NO}_3^-\text{-N}$ from $\text{NH}_4^+\text{-N}$ in soil leachate was due to its complicated mechanism, this indicated that $\text{NO}_3^-\text{-N}$ in soil leachate mainly occurred in drained soil situation. This coincided with other studies' results (Lu et al. 2006; Wang et al. 2009). But effect of water regime and fertilizer on variation of $\text{NO}_3^-\text{-N}$ in percolation was still complicated due to intricate factors which could influence the process of nitrification and denitrification (Wang et al. 2009).

Vertical distribution of N leaching

A pronounced vertical distribution of N leaching was investigated. The concentration of $\text{NH}_4^+\text{-N}$ decreased with the increasing of sampling depth in range of 0-100 cm. Average concentration of $\text{NH}_4^+\text{-N}$ in 20-30 cm during two years was: 0.78 mg L^{-1} , and for 50-60 cm and 80-90 cm, it was 0.61 mg L^{-1} and 0.47 mg L^{-1} respectively. Significant difference was observed in the vertical distribution of $\text{NH}_4^+\text{-N}$ concentrations ($p < 0.05$) except NPKS in early rice growing season 2006, NPK in late rice growing season 2006, and NPK in both seasons of 2007. However, the concentration of $\text{NO}_3^-\text{-N}$ exhibited an increased trend in first 60 cm and then decreased from 60 cm to 100 cm. For the three sampling depths, the average concentration of nitrate-N was 1.63 mg L^{-1} , 3.56 mg L^{-1} and 2.62 mg L^{-1} respectively, while significant difference was found only in NPKS and CK of 2006 ($p < 0.05$). Many studies found the decline tendency of $\text{NH}_4^+\text{-N}$ with the depth of soil aquifers in rice field (Wang et al. 1997; Yin et al. 2005), its coincide with our results. Whereas, the results of vertical distribution of $\text{NO}_3^-\text{-N}$ in percolation observed were different from some other studies. Wang et al. (1997) and Yin et al. (2005) found the concentration of $\text{NO}_3^-\text{-N}$ in soil solution increased with the incremental depth in the range of 0-100 cm, but by Wang et al. (2009) considered that the vertical distribution of concentration of $\text{NO}_3^-\text{-N}$ depended on water regime and fertilization applied on rice production.

Impact of long-term fertilization on N leaching

Formation and amount of fertilizer has a pronounced influence on N leaching in rice field. By statistical analysis, concentrations of N leaching of CK in all the aquifers was relatively low, and

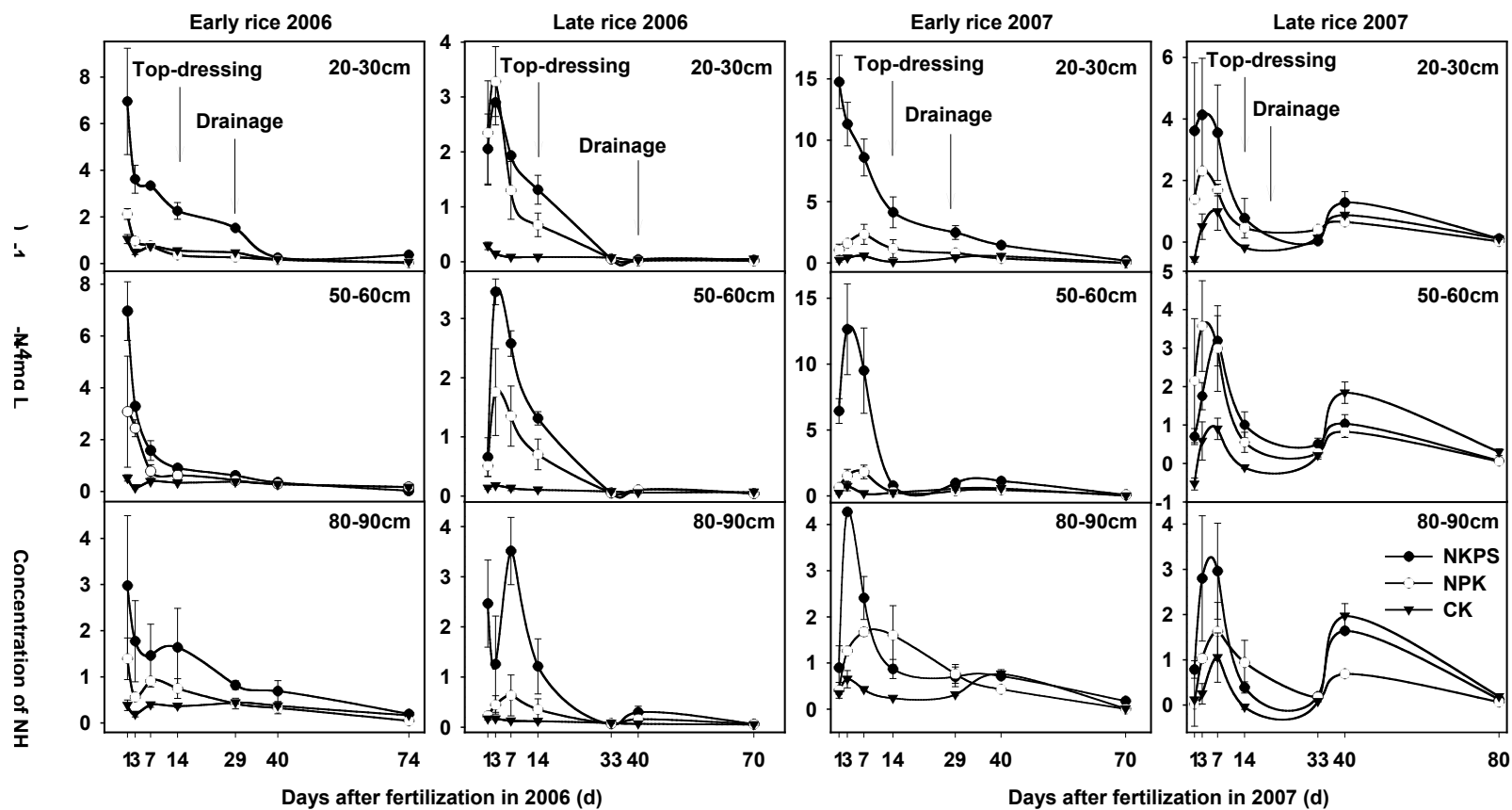


Figure 3. Seasonal variation of concentration of $\text{NH}_4^+\text{-N}$ in soil solution of different depths

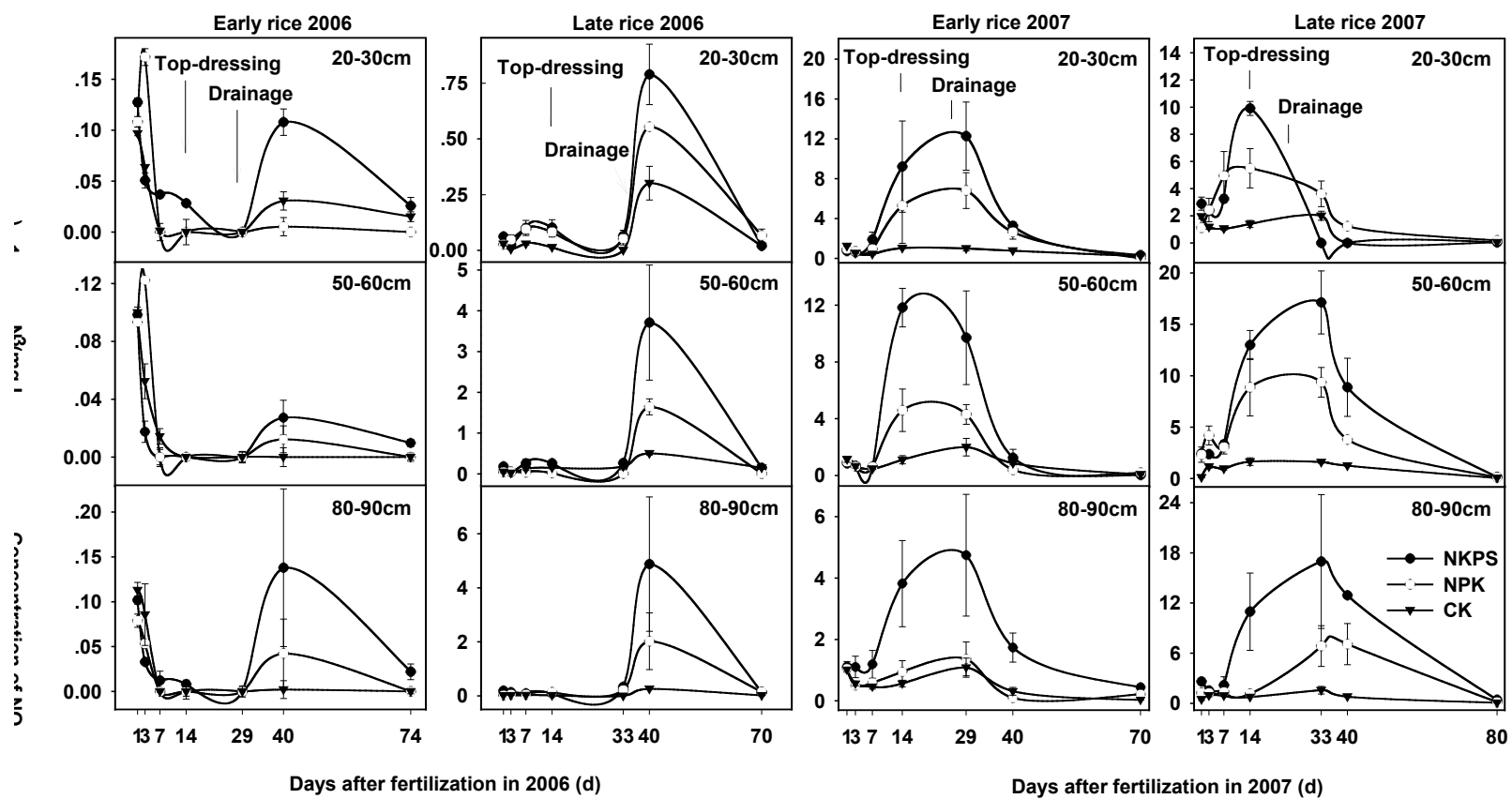


Figure 4. Seasonal variation of concentration of NO_3^- -N in soil solution of different depths

significantly less than NPKS and NPK ($p < 0.05$). The mean value of concentration of $\text{NH}_4^+\text{-N}$ in three depths' leachate of soil solutions of NPKS, NPK and CK in two years was 0.82 mg L^{-1} , 0.43 mg L^{-1} and 0.32 mg L^{-1} respectively. For $\text{NO}_3^-\text{-N}$, the average value for three treatments was 3.11 mg L^{-1} and 2.10 mg L^{-1} and 0.48 mg L^{-1} respectively. While this regularity varied by different rice growing seasons. For early rice growing season during two years, concentration of $\text{NH}_4^+\text{-N}$ of NPKS was 203.11 % and 240.91 % greater than NPK and CK respectively; and for $\text{NO}_3^-\text{-N}$ the increments for two treatments were 117.72 % and 395.53 % respectively. But for late rice growing season, the average value of concentration of $\text{NH}_4^+\text{-N}$ of NPKS was 18.44 % less than NPK while as still 41.23 % greater than CK; for $\text{NO}_3^-\text{-N}$, the increments for two treatments were 25.56 % and 672.95 % respectively. It indicated that long-term chemical nitrogenous fertilizer incorporated with organic fertilizer (rice straw) led to more N leaching in rice field than only chemical fertilizer was applied and even more than the treatment with zero fertilizer, but this regularity depended by different rice growing season, such as our results. Its no doubt that the excessive N provided by rice straw can be involved into and strengthen the evolution of downward motion of $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ in soil leachate. Tong et al. (1997) also found organic manure could cause greater N leaching. In fact, many studies also considered the N losses in rice field was highly related with increasing applied amount of chemical N fertilizer (Jin and Yang 2004; Wang et al. 1997; Yin et al. 2005; Kurosawa et al. 2007).

For the N leaching losses in early rice growing season, $\text{NO}_3^-\text{-N}$ contributed 64.10 %, the other 35.90 % came from $\text{NH}_4^+\text{-N}$. Moreover, for the late rice growing season, $\text{NO}_3^-\text{-N}$ and $\text{NH}_4^+\text{-N}$ caused 84.70 % and 15.30 % of N leaching losses respectively. It indicated that $\text{NO}_3^-\text{-N}$ leaching losses were the main source of N pollution in soil percolation of rice field in Hunan, China. Consequently, $\text{NO}_3^-\text{-N}$ is the primary form of N leaching in rice field instead of $\text{NH}_4^+\text{-N}$, the two years' average concentration of $\text{NO}_3^-\text{-N}$ is 2.61 mg L^{-1} , while for $\text{NH}_4^+\text{-N}$, it was 0.62 mg L^{-1} . Many studies found the nitrate-N is the main form of N leaching in percolation water of rice field (Wang et al. 1997). While $\text{NH}_4^+\text{-N}$ was found is the main form of N leaching in a Japan rice field, and the concentration of $\text{NO}_3^-\text{-N}$ was extremely low based on a long-term fertilization experiment (Luo et al, 2010), however the same author provided a different conclusion in a previous study, which pointed that in northeastern China, N leaching from rice soils were mainly contributed by $\text{NO}_3^-\text{-N}$ and $\text{NH}_4^+\text{-N}$ accounted only small part of them (Luo et al 2000). These opposite phenomenon maybe due to the variation of spatial distribution of rice field.

Leaching losses in rice field

Relatively high total N leaching losses was found in two years' study, NPKS led greater N leaching losses than NPK and CK, and N leaching losses in later rice growing season was greater than early rice growing season (Table 2). Table 2 showed that N leaching losses by ammonium and nitrate were comparatively high (up to $31.35 \text{ kg N ha}^{-1}$ and 17.40 % of applied N for NPKS) in continuous submerged rice field. In another rice field, much higher N leaching rate was reported by Takeda et al. (1991), in which the N leaching losses rate was $95\text{-}122 \text{ kg N ha}^{-1}$. However, different conclusion provided by Luo et al. (2010), who found a relatively lower N leaching losses from the treatment of

composted rice straw plus soybean cake ($0.58 \text{ kg N ha}^{-1}$) than the treatment of ammonium sulphate ($2.41 \text{ kg N ha}^{-1}$) from a long-term fertilization rice field of Japan and only less than 3 % of applied N was found in soil solution. Panda et al. (1989) pointed that this low N leaching rate was attributed to a low percolation rate, which was due to a high groundwater table and small volumes of irrigation water was used (Luo et al 2010). Another point of view provided by Kyaw et al. (2005) believed that the low leaching rate should be attributed to restriction of surface outflow, which was found caused relatively lower N leaching rate (6.0 kg N ha^{-1}) compared to the experiment with no control on surface outflow ($42\text{-}48 \text{ kg N ha}^{-1}$) (Takeda et al. 1991). All of these studies considered irrigation as an important factor to regulate the N leaching rate in rice field, because the irrigation water used in the rice field may be contain a higher N concentration (Kyaw et al. 2005; Hidaka 1994), this could explain the high N leaching rate showed in our study. In fact, N leaching from rice paddy soil showed a great variation around the world, in humid areas N leaching was found account to up to 50 % of the applied N (Tisdale et al. 1993). All of the differences discovered above indicated that there was great variation of N leaching from rice field due to not only different type and application rate of N fertilizers but also heterogeneously distribution of geographical environment (Luo et al, 2010) and the different growing stage of rice cultivars (Cao et al. 2005) and variant rice species planted as in our study.

Table 2 Total N Losses (Kg N Ha^{-1}) and Loss Ratio (%) in Three Treatments

	Early rice growing season		Later rice growing season	
	N*	ratio	N	ratio
NPKS	22.67	15.11	31.35	17.41
NPK	9.35	6.23	26.10	14.50
CK	5.33	3.55	5.58	3.10

*Total N losses include summary of three aquifers' ammonium-N and nitrate-N; leaching ratio was calculated via applied N divided by N leaching amount in that season. The value was the two years' average value

Conclusions

The employed porous suction pipes method performed well in double rice field for N leaching investigation. As affected by long-term fertilization, there was a significant temporal and vertical distribution of N leaching in soil percolation of rice field. Generally speaking, $\text{NH}_4^+\text{-N}$ in soil leachate mainly occurs in submerged situation before MSD, while $\text{NO}_3^-\text{-N}$ could primarily be found in situation with relatively lower soil water content. Moreover, a pronounced vertical trend of N leaching was also found in 0-100 cm of soil leachate in rice field. However, the concentration of $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ exhibited a different distribution with the increase of depth of sampling.

Owing to long-term additional organic fertilizer, NPKS caused greater N leaching loss than NPK and CK in irrigated double rice field. The nitrate-N was the primary formation of N leaching in study area. However, the potential hazards of nitrate leaching was low due to most concentration of $\text{NO}_3^-\text{-N}$ observed in soil percolation were less than 10 mg

L⁻¹ (Fig 3 and Fig 4), which was the maximum contaminant level of NO₃⁻-N in groundwater (US Environmental Protection Agency 2009).

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